

9-26-00

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## UTILITY PATENT APPLICATION TRANSMITTAL

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Box Patent Application  
Washington, D.C. 20231

Attorney Docket No. 205701

First Named Inventor Igarashi

Express Mail No. EL303750267US

## APPLICATION ELEMENTS

1. ☒ Utility Transmittal Form
2. ☒ Specification (including claims and abstract) [Total Pages 10]
3. ☐ Drawings [Total Sheets ]
4. ☒ Combined Declaration and Power of Attorney [Total Pages 4]
  - a. ☒ Newly executed
  - b. ☐ Copy from prior application
  - i. ☐ **[Note Box 5 below]** Deletion of Inventor(s) Signed statement attached deleting inventor(s) named in the prior application
5. ☐ Incorporation by Reference: The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4b is considered as being part of the disclosure of the accompanying application and is hereby incorporated by reference therein.
6. ☐ Microfiche Computer Program
7. ☐ Nucleotide and/or Amino Acid Sequence Submission
  - a. ☐ Computer Readable Copy
  - b. ☐ Paper Copy
  - c. ☐ Statement verifying above copies

## ACCOMPANYING APPLICATION PARTS

8. ☒ Assignment Papers (cover sheet and document(s))
9. ☐ Power of Attorney
10. ☐ English Translation Document (if applicable)
11. ☐ Information Disclosure Statement (IDS)
  - ☐ Form PTO-1449
  - ☐ Copies of References
12. ☐ Preliminary Amendment
13. ☒ Return Receipt Postcard (Should be specifically itemized)
14. ☐ Small Entity Statement(s)
  - ☐ Enclosed
  - ☐ Statement filed in prior application; status still proper and desired
15. ☒ Certified Copy of Priority Document(s)
16. ☐ Other:

17. **If a CONTINUING APPLICATION**, check appropriate box and supply the requisite information in (a) and (b) below:
- (a) ☐ Continuation ☐ Divisional ☐ Continuation-in-part of prior application Serial No. \_\_\_\_\_  
Prior application information: Examiner \_\_\_\_\_; Group Art Unit \_\_\_\_\_
- (b) Preliminary Amendment: Relate Back - 35 USC §120. The Commissioner is requested to amend the specification by inserting the following sentence before the first line:  
"This is a ☐ continuation ☐ divisional of copending application(s)  
☐ Application No. \_\_\_\_\_, filed on \_\_\_\_\_,  
☐ International Application, filed on \_\_\_\_\_, and which designates the U.S."

## APPLICATION FEES

BASIC FEE				\$690.00
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total Claims	14 -20=	0	x \$18.00	\$0
Independent Claims	2 -3=	0	x \$78.00	\$0
<input checked="" type="checkbox"/> Multiple Dependent Claim if applicable				+\$260.00 \$260.00
Total of above calculations =				\$950.00
Reduction by 50% for filing by small entity =				\$( )
<input checked="" type="checkbox"/> Assignment fee if applicable				+ \$40.00 \$40.00
TOTAL =				\$990.00

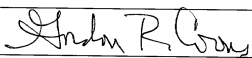
## UTILITY PATENT APPLICATION TRANSMITTAL

Attorney Docket No. 205701

19. ☐ Please charge my Deposit Account No. 12-1216 in the amount of \$
20. ☒ A check in the amount of \$990.00 is enclosed.
21. The Commissioner is hereby authorized to credit overpayments or charge any additional fees of the following types to Deposit Account No. 12-1216:
- a. ☒ Fees required under 37 CFR §1.16.
- b. ☒ Fees required under 37 CFR §1.17.
22. ☐ The Commissioner is hereby generally authorized under 37 CFR §1.136(a)(3) to treat any future reply in this or any related application filed pursuant to 37 CFR §1.53 requiring an extension of time as incorporating a request therefor, and the Commissioner is hereby specifically authorized to charge Deposit Account No. 12-1216 for any fee that may be due in connection with such a request for an extension of time.

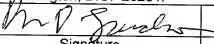
## 23. CORRESPONDENCE ADDRESS

Gordon R. Coons, Registration No. 20821  
Leydig, Voit & Mayer, Ltd.  
Two Prudential Plaza, Suite 4900  
180 North Stetson  
Chicago, Illinois 60601-6780  
Telephone: (312) 616-5600  
Facsimile: (312) 616-5700

Name	Gordon R. Coons
Signature	
Date	September 25, 2000

## Certificate of Mailing Under 37 CFR §1.10

I hereby certify that this Utility Patent Application Transmittal and all accompanying documents are being deposited with the United States Postal Service "Express Mail Post Office To Addressee" Service under 37 CFR §1.10 on the date indicated below and is addressed to: Commissioner of Patents and Trademarks, Box Patent Application, Washington, D.C. 20231.

Brian D. Sandstrom		September 25, 2000
Name of Person Signing	Signature	Date

UTILITY (Rev. 6/29/2000)

## METHOD FOR PRODUCING HYDROCYANIC ACID SYNTHESIS CATALYST

### FIELD OF THE INVENTION

The present invention, relates to a method for producing a catalyst that is  
5 involved in the reaction for the production of hydrocyanic acid and water by  
thermal decomposition of formamide. It more specifically relates to a catalyst  
production method which may be put into practice commercially using an  
extremely simple process which does not make use of corrosive liquid reagents or  
the like during the preparation of a catalyst and which does not require further  
10 treatment after preparation.

### BACKGROUND OF THE INVENTION

Methods for the synthesis of hydrocyanic acid by the selective thermal  
decomposition of formamide using a catalyst made from a metal or a metal oxide  
15 have long been known. With regard to the catalyst used, many are known in the  
prior art, and, for example, cyanide is synthesized from formamide using a pipe  
catalyst prepared by impregnating aluminum into the surface of a reaction pipe  
using a melted aluminum bath to create an iron - aluminum alloy as is reported in  
Japanese Patent 1960-9182. Additionally Japanese Patent 1958-6229 discloses a  
20 method for preparing catalyst by treating a saturated solution of potassium  
bichromate in concentrated sulfuric acid, a solution of potassium permanganate in  
concentrated sulfuric acid, aqueous hydrogen peroxide and the like. To carry out  
these methods on an industrial scale, it is necessary to prepare several specialized  
items of equipment for conducting various treatments.

The method according to Japanese Patent 1960-9182 for preparing an  
oxidized layer where a steel pipe is used in the reactor requires a very large  
investment for the application of industrial equipment made of multiple pipe  
reactors by methods for preparing reaction pipes. In addition where an oxidative  
reagent is used in the same manner as in Japanese Patent 1958-6229, it is  
30 necessary to add a separate piece of equipment to prepare the reagents, and reagent  
left in the reaction equipment for unspecified periods of time can cause corrosion

trouble. Moreover, during and after treatment with the reagent, it is necessary to conduct and administer complicated and difficult procedures to recycle the reagent during and after treatment.

Moreover because the catalyst layer prepared by the use of reagents  
5 exhibits poor adhesion, and because it cannot endure long continuous use, this method exhibits the defect of almost all of the catalyst separating and falling away. Without even taking into account the loss of catalyst activity caused by the peeling off of the oxide layer, this results in the additional bad effect of contamination of the system by the metal oxides that have peeled off. The inventors of the present  
10 invention achieved the present invention as a result of intensive and purposeful research to discover a method for solving these problems.

### SUMMARY OF THE INVENTION

The object of the present invention is to offer a method for producing  
15 catalyst for the synthesis of hydrocyanic acid by thermal decomposition of formamide which exhibits both high activity and stability over a long period of time.

### DETAILED DESCRIPTION OF THE INVENTION

20 The inventors of the present invention conducted various investigations to arrive at the discovery of a method of treatment by a gas as a method for preparing an oxide layer without carrying out a reagent treatment on the surface of the pipe. The results were that it was relatively easy to prepare an oxide layer without carrying out a reagent treatment on the surface of the pipe. However, preparation  
25 of a layer with catalyst performance and strong adhesion were not easily accomplished. The results of further investigations were that by treatment of the metal surface by means of a method that satisfied certain specific conditions, it was observed that it was possible to create an oxide layer possessing high catalyst performance and strong adhesion. More specifically, by sequential treatment  
30 involving controlled oxidative and reductive gas, an invention in which the preparation of an oxide layer that possesses catalyst performance and moreover

good adhesive strength were observed was obtained. That is to say, the present invention relates to a method for the preparation of a catalyst for the synthesis of hydrocyanic acid characterized by sequential exposure of an iron surface to an oxidative gas and a reductive gas.

5 As for the iron which is a raw material used in the method for preparing the catalyst of the present invention, illustrative preferred examples include pig iron containing about 2% by weight or more of carbon, steel containing carbon less than about 2% by weight and additionally, steels characterized by the addition to these carbon steels of alloying elements other than carbon may be used. However,  
10 as may be appreciated, any source of iron may be used that can be utilized to carry out the desired synthesis. Many such sources are known.

The form or shape of the iron used in the method for producing a catalyst of the present invention varies according to the way in which the catalyst is used. In the case where the catalyst is packed in a column, a particulate or powdered  
15 form is preferred and the surface of these particulate or powdered irons are made into an oxide layer. When the catalyst is used as a reaction pipe, from the stand point of longevity, the use of steel is preferred as the iron source and the internal surface of the steel pipe is made into an oxide layer.

When the surface of the iron of the present invention is exposed to an  
20 oxidative atmosphere and a reductive atmosphere, the conditions are preferably as follows. The oxidative atmosphere is possible to achieve by covering the iron surface with a gas containing between 5% and 30% by volume of oxygen. An easy and preferred method is to cover the surface with air. The temperature of treatment during exposure to an oxidative atmosphere is between 300°C and  
25 650°C, the treatment pressure is between 6 kPa and 150 kPa, the space velocity (referred to below as "SV") is from 10 to 1500 h<sup>-1</sup> and a time for one cycle of this phase of treatment of between 5 and 300 minutes is preferred.

An illustrative example of a reductive atmosphere is to cover the iron surface with a gas containing 1% and 99% by volume of hydrogen, preferably between 2%  
30 and 20% by volume of hydrogen. The oxygen content contained in the gas should be as little as possible. Covering the surface with a gas composed of hydrogen and

steam, or a gas composed of hydrogen and nitrogen and steam are preferred. The temperature of treatment during exposure to an oxidative atmosphere is preferably between 300°C and 650°C, the treatment pressure is between 6 kPa and 150 kPa, an SV of from 10 to 1500 h<sup>-1</sup> and a time for one cycle of this phase of treatment of  
5 between 10 and 300 minutes. Repetitive cycles of respective exposure to an oxidative atmosphere and exposure to a reductive atmosphere are preferred. It is thus preferred that at least two cycles be used; and, indeed, 10 to 40 or more cycles can be desirably utilized. Where repeated cycles of respective exposure to an oxidative atmosphere and exposure to a reductive atmosphere are both repeated, it does not matter which  
10 atmosphere is introduced first.

Where hydrocyanic acid is produced by the thermal decomposition of formamide using the catalyst of the present invention, it is preferred that the gas phase decomposition reaction be carried out continuously, and the formamide which constitutes the raw material is supplied to the reactor as a gas. In this case,  
15 illustrative useful reaction conditions include a reaction temperature of from 300°C to 600°C, a reaction pressure of from 6 to 150 kPa, an SV of from 50 to 1500 h<sup>-1</sup> for between 60 and 3000 minutes.

It will generally be desired to regenerate the catalyst when the activity has declined below the desired level. The activity can be determined in reference to  
20 the conversion and/or yield. Any other parameter or parameters may, of course, be utilized as is desired. Regeneration is carried out with gas containing from 5% to 30% by volume of oxygen and, preferably, in air, having a temperature of between 300° to 600°C, a pressure of from 6 to 150 kPa, an SV of from 10 to 1500 h<sup>-1</sup> for a period of between 10 and 500 minutes. It is possible to make  
25 hydrocyanic acid from formamide by repeating the aforementioned reaction and regeneration technique.

By using the method of the present invention, it is possible to minimize the peripheral equipment needed for the preparation of a hydrocyanic acid synthesis catalyst without the disposal of used reagent or separated oxide layer, the  
30 economic effects of which are favorable and large.

The catalyst produced by the present invention, in comparison to prior catalysts, is characterized by smaller, more compact crystals, free of any

contaminants, such as trapped liquid residue or the like. These characteristics, and particularly the crystal size contribute to an enhanced initial effectiveness and stability. Thus, use of the catalyst of this invention allows superior output of hydrocyanic acid, without fouling or the like, at the initial stages of operation.

The practice examples that follow are for the purpose of explaining the Present Invention by illustrating some of the practical embodiments within its scope, but are in no way intended to limit the scope of the present invention or the ways in which it may be practiced.

### Example 1

A steel pipe having an internal diameter of 10.9 mm and length of 448 mm was selected as the material for the reaction pipe. After conducting treatment under an oxidative atmosphere for 50 minutes by supplying the inside of the steel pipe with air at SV  $80 \text{ h}^{-1}$  at a pressure of 13.3 k Pa and a temperature of  $400^{\circ}\text{C}$ , under the same conditions the gas was switched to a gas composed of a hydrogen, nitrogen and steam in a molar ratio of 6:17:77 at SV  $350 \text{ h}^{-1}$  and this reductive treatment was continued for 180 minutes. The aforementioned oxidative and reductive treatments were alternatively repeated 10 times each and a hydrocyanic acid synthesis catalyst was obtained by the creation of an oxide layer inside the pipe.

Subsequently, using the catalyst thus obtained, hydrocyanic acid was produced by thermal decomposition of formamide. Formamide gas was passed over the catalyst inner surface of the pipe at SV  $350 \text{ h}^{-1}$ . The reaction temperature was  $400^{\circ}\text{C}$  and the reaction pressure was 13.3 kPa, and after conducting the reaction for 60 minute from start to obtain a formamide in a conversion ratio of 99.6 mole% and a hydrocyanic acid yield of 94.6 mole%.

### Example 2

A steel pipe having an internal diameter of 10.9 mm and length of 448 mm was selected as the material for the reaction pipe. After conducting treatment under an oxidative atmosphere for 5 minutes by supplying the inside of the steel

pipe with air at SV 260 h<sup>-1</sup> at a pressure of 13.3 kPa and a temperature of 400°C, under the same conditions the gas was switched to a gas composed of a hydrogen, nitrogen and steam in a molar ratio of 6:17:77 at SV 480 h<sup>-1</sup> and this reductive treatment was continued for 45 minutes. The aforementioned oxidative and reductive treatments were alternatively repeated 40 times each and a hydrocyanic acid synthesis catalyst was obtained by the creation of an oxide layer inside the pipe.

Subsequently, using the catalyst thus obtained, hydrocyanic acid was produced by thermal decomposition of formamide under the same conditions as in Example 1. The reaction was carried out for 60 minutes to obtain a formamide in a conversion ratio of 99.2 mole% and a hydrocyanic acid yield of 94.2 mole%.

### Comparative Example 1

Except for the use of a steel pipe having an internal diameter of 10.9 mm and length of 448 mm which had not been subjected to oxidation and reduction treatment as the reaction pipe, a thermal decomposition of formamide was carried out under the same conditions as in Example 1. The reaction was continued for 60 minutes from the start of the reaction and a formamide conversion ratio of 36.5 mole% and a hydrocyanic acid yield of 34.7 mole% were obtained.

### Example 3

In order to confirm the strength of adhesion of the catalyst made according to Example 1, a continuous process for 7 days was carried out consisting of alternate formamide thermal decomposition reaction and catalyst regeneration carried out using air to counter the decline in catalyst performance due to adhesion of carbon materials on the catalyst's surface. To measure the volume of detached catalyst that came about during this procedure, the pipe was disconnected. There was no accumulated iron oxide in the mouth of the pipe. The thermal decomposition of formamide was carried out for 7 hours at a time at a reaction temperature of 400°C, a reaction pressure of 13.3 kPa while formamide gas was supplied to the inside of the reaction pipe at an SV of 700 h<sup>-1</sup> and the catalyst



regeneration was carried out by passing air through the inside of the reaction pipe at an SV of  $300 \text{ h}^{-1}$ , a treatment temperature of  $400^\circ\text{C}$  and a reaction pressure of  $13.3 \text{ kPa}$  for 1 hour at a time.

5

### Comparative Example 2

- Using a steel pipe the same as was used in Example 1, an oxidative layer was formed by oxidative reagents. After impregnating for 48 hours using the reagent, a saturated solution of iron formate in aqueous formic acid, and then was heated under a flow of air at  $400^\circ\text{C}$  for one hour. In the reaction pipe, a  $0.5 \text{ mm}$  thick accumulation of iron oxide layer had formed. Next, using this reaction pipe, the same operation as in Example 3 which included alternate thermal decomposition of formamide and catalyst regeneration was conducted for 7 days continuously. From the mouth of the reaction pipe when it was opened for investigation was taken an accumulation of  $1.3$  grams composed of iron oxide. This accumulated amount was equal to the amount of the iron oxide layer on the surface of the reaction pipe before the reaction was started and as a result it was observed that almost all of the layer had fallen off.

## WE CLAIM:

1. A method for producing hydrocyanic acid synthesis catalyst characterized by sequential exposure of an iron surface to oxidative and reductive atmospheres.
2. A method for producing hydrocyanic acid synthesis catalyst according to claim 1 wherein the inner surface of an iron pipe is sequentially exposed to oxidative and reductive atmospheres.
3. A method for producing hydrocyanic acid synthesis catalyst according to claim 1 or claim 2 wherein the cycle of exposure to oxidative and reductive atmospheres is carried out more than once.
4. The method of claim 1 wherein the oxidative atmosphere is a gas containing from about 5% to 30% by volume of oxygen.
5. The method of claim 1 wherein the reductive atmosphere is a gas containing between 1% and 99% by volume of hydrogen.
6. The method of claim 1 wherein the oxidative atmosphere is a gas containing from about 5% to 30% by volume of oxygen and the reductive atmosphere is a gas containing between 1% and 99% by volume of hydrogen.
7. The method of claim 1 wherein the exposure of the iron surface to the oxidative atmosphere is conducted under the conditions that the temperature is in the range of about 300°C to 650°C, the pressure is in the range of from about 6 kPa to 150 kPa, a space velocity of from about 10 to 150h<sup>-1</sup> and the time is from about 5 to 300 minutes.
8. The method of claim 1 wherein the exposure of the iron surface to the reductive atmosphere is conducted under the conditions that the temperature is

in the range of about 300°C to 650°C, the pressure is in the range of from about 6 kPa to 150 kPa, a space velocity of from about 10 to 150h<sup>-1</sup> and the time is from about 5 to 300 minutes.

9. The method of claim 1 wherein the exposure of the iron surface to the oxidative and reductive atmospheres is conducted under the conditions that the temperature is in the range of about 300°C to 650°C, the pressure is in the range of from about 6 kPa to 150 kPa, a space velocity of from about 10 to 150h<sup>-1</sup> and the time is from about 5 to 300 minutes.

10. A hydrocyanic acid synthesis catalyst characterized by the presence of a surface having an oxide layer that has been prepared by sequential exposure of an iron surface to oxidative and reductive atmospheres.

11. A method for making hydrocyanic acid which comprises exposing formamide to a catalyst made by the method of claim 1 under reaction conditions to provide hydrocyanic acid.

12. The method of claim 11 wherein the formamide is a gas, and the reaction conditions include a temperature in the range of about 300°C to 600°C, a pressure of from about 6 to 150 kPa, a space velocity of from about 50 to 1500h<sup>-1</sup> for between about 60 to 3000 minutes.

**ABSTRACT OF THE DISCLOSURE**

A method for producing a hydrocyanic acid synthesis catalyst is disclosed which comprises sequential exposure of an iron surface, such as, for example, pig iron, to sequential oxidative and reductive atmospheres, preferably using repetitive cycles, the oxidative atmosphere preferably containing 5% to 3% by volume oxygen and the reductive atmosphere preferably containing 1% to 99% hydrogen.

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COMBINED DECLARATION AND POWER OF ATTORNEY

As below named inventor, I hereby declare that

This declaration is of the following type:

- ☒ original ☐ design ☐ supplemental  
☐ national stage of PCT  
☐ divisional ☐ continuation ☐ continuation-in-part

My residence, post office address, and citizenship are as stated below next to my name. I believe I am the original, first, and sole inventor (*if only one name is listed below*) or an original, first, and joint inventor (*if plural names are listed below*) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METHOD FOR PRODUCING HYDROCYANIC ACID SYNTHESIS  
CATALYST

the specification of which:

- ☒ is attached hereto.  
☐ was filed on \_\_\_\_\_ as Application No. \_\_\_\_\_ and was amended on \_\_\_\_\_ (*if applicable*).  
☐ was filed by Express Mail No. \_\_\_\_\_ as Application No. not known yet, and was amended on \_\_\_\_\_ (*if applicable*).  
☐ was described and claimed in PCT International Application No. \_\_\_\_\_ filed on \_\_\_\_\_ and as amended pursuant to PCT Article 19 on \_\_\_\_\_ (*if any*).

I state that I have reviewed and understand the contents of the above-identified specification, including the claim(s), as amended by any amendment referred to above.

I acknowledge the duty to disclose information that is material to the patentability of this application in accordance with 37 C.F.R. § 1.56.

I claim foreign priority benefits under 35 U.S.C. § 119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent, utility model, design registration, or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

PRIOR FOREIGN PATENT, UTILITY MODEL, AND DESIGN REGISTRATION APPLICATIONS						
COUNTRY	APPLICATION	DATE OF FILING (day,month,year)	PRIORITY CLAIMED UNDER 35 U.S.C. § 119			
Japan	11-272247	27 September 1999	X	YES		NO
				YES		NO
				YES		NO

I claim the benefit pursuant to 35 U.S.C. § 119(e) of the following United States provisional application(s):

PRIOR U.S. PROVISIONAL APPLICATIONS BENEFIT CLAIMED UNDER 35 U.S.C. 119(e)	
APPLICATION NO.	DATE OF FILING (day,month,year)

I claim the benefit pursuant to 35 U.S.C. § 120 of any United States application(s) or PCT international application(s) designating the United States of America listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose material information as defined in 37 C.F.R. § 1.56 effective between the filing date of the prior application(s) and the national or PCT international filing date of this application.

PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL PATENT APPLICATIONS DESIGNATING THE U.S. FOR BENEFIT UNDER 35 U.S.C. 120					
U.S. APPLICATIONS			Status (check one)		
APPLICATION NO.	U.S. FILING DATE	PATENTED	PENDING	ABANDONED	
1. 0 /					
2. 0 /					
3. 0 /					
PCT APPLICATIONS DESIGNATING THE U.S.			Status (check one)		
PCT APPLICATION NO.	PCT FILING DATE (day,month,year)	U.S. APPLN. NOS. ASSIGNED (if any)	PATENTED	PENDING	ABANDONED
4.					
5.					
6.					

DETAILS OF FOREIGN APPLICATIONS FROM WHICH PRIORITY CLAIMED UNDER 35 U.S.C. §119 FOR ABOVE LISTED U.S./PCT APPLICATIONS				
ABOVE APPLN. NO.	COUNTRY	APPLICATION NO.	DATE OF FILING (day,month,year)	DATE OF ISSUE (day,month,year)
1.				
2.				
3.				
4.				
5.				
6.				

As a named inventor, I hereby appoint the following attorneys to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

In re Appln. of Igarashi  
Attorney Docket No. 205701

Berton Scott Sheppard, Reg. 20922  
James B. Muskall, Reg. 22797  
Dennis R. Schlemmer, Reg. 24703  
Gordon R. Coons, Reg. 20821  
John E. Rosenquist, Reg. 26356  
John W. Kozak, Reg. 25117  
Charles S. Olakovic, Reg. 27583  
Mark E. Phelps, Reg. 28461  
H. Michael Hartman, Reg. 28423  
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Charles H. Mottier, Reg. 30874  
John Kilyk, Jr., Reg. 30763  
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James D. Zalewa, Reg. 27848  
John M. Belz, Reg. 30359  
Brett A. Hesterberg, Reg. 31837  
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Paul J. Korniczky, Reg. 32849

Pamela J. Ruschau, Reg. 34242  
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Y. Kurt Chang, Reg. 41397  
Gregory C. Bays, Reg. 40505  
Carol Larcher, Reg. 52243  
Steven H. Sider, Reg. 42154  
M. Daniel Hefner, Reg. 41826  
Thomas A. Belush, Reg. 37090

Kenneth P. Spina, Reg. 43927  
Siegmund Y. Gutman, Reg. 46304  
Andrew J. Heinisch, Reg. 43666  
Jeffery J. Makeover, Reg. 37390  
Salim A. Hasan, Reg. 38175  
Richard A. Wulff, Reg. 42238  
Phillip M. Pippenger, Reg. 46055  
Jamison E. Lynch, Reg. 41168  
Vladan M. Vasiljevic, Reg. 45177  
Rattan Nath, Reg. 43827  
Robert M. Gould, Reg. 43642  
Len Smith, Reg. 43139  
Kevin L. Wingate, Reg. 38662  
David J. Schodin, Reg. 41294  
Heather R. Kissling, Reg. 45790  
Paul L. Ahern, Reg. 17020  
Theodore W. Anderson, Reg. 17035  
Noel I. Smith, Reg. 18698

I further direct that correspondence concerning this application be directed to LEYDIG, VOIT & MAYER, LTD., Two Prudential Plaza, Suite 4900, 180 North Stetson, Chicago, Illinois 60601-6780, Telephone (312) 616-5600.

I declare that all statements made herein of my own knowledge are true, that all statements made on information and belief are believed to be true, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor: TAKESHI IGARASHI

Inventor's signature Takeshi Igarashi

Date September 11, 2000

Country of Citizenship: Japan

Residence: Niigata-ken, Japan

Post Office Address: c/o Nigata Factory, Mitsubishi Gas Chemical Company, Inc., 3500 Matsuhamma-cho, Niigata-shi, Niigata-ken, Japan

Full name of second joint inventor, if any: SUSUMU NAITO

Inventor's signature Susumu Naito

Date September 11, 2000

Country of Citizenship: Japan

Residence: Niigata-ken, Japan

Post Office Address: c/o Nigata Factory, Mitsubishi Gas Chemical Company, Inc., 3500 Matsuhamma-cho, Niigata-shi, Niigata-ken, Japan

In re Appln. of Igarashi  
Attorney Docket No. 205701

Full name of **third joint inventor**, if any: TOHRU NAKAMURA

Inventor's signature Tohru Nakamura

Date September 11, 2000

Country of Citizenship: Japan

Residence: Niigata-ken, Japan

Post Office Address: c/o Nigata Factory, Mitsubishi Gas Chemical Company, Inc., 3500 Matsuhama-cho,  
Niigata-shi, Niigata-ken, Japan